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BUDAPEST

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ABSTRACT

The dispersion relation of surface plasma oscillations of the Au-vacuum interface was determined from the light emission of Al-Al_2O_3-Au tunnel diodes. The comparison of results with previous data obtained on Ag indicate that the mechanism of light generation is qualitatively the same for all MOM structures if the anode is one of the noble metals.

АННОТАЦИЯ

Дисперсионный закон ПЭВ на разделе золото-вакуум был определён в Al-Al_2O_3-Au туннельных диодах измеряя спектральное и угловое распределения излучаемого света. Сравнение результатов измерений с данными, полученными раньше для Ag показало, что механизм генерации света является качественно тождественным во всех МОМ структурах с анодом из драгоценных металлов.

KIVONAT

Al-Al_2O_3-Au MOM tunnel diodákból kilépő fény spektrális és szögeloszlásának mérésével meghatároztuk az arany-vákuum felülethez tartozó felületi plazmonok diszperziós összefüggését. A kisérleti eredményeket az ezüstre vonatkozó korábbi adatokhoz hasonlitva megállapitható, hogy mindazokban a MOM szerkezetekben, ahol az anód nemes-fém, a fénykibocsátás mechanizmusa lényegében azonos.

It has been shown experimentally that the $\omega(k)$ dispersion relation of surface plasma oscillations /SPO/ of metal-vacuum or metal-insulator interfaces can be determined by measuring the angular and spectral distribution of light, emitted by metal oxide-metal /MOM/tunnel diodes made on periodically corrugated substrates [1,2]. This dispersion relation, however, differs from that calculated for an ideally smooth metal surface, although the differences are not too large [2,3]. The dominant anomaly can be described as a shift of plasmon resonances towards larger k-values, explained by the scattering of plasmons on statistical irregularities of the metal-vacuum interface. With increasing depth of grooves of the gratings /h/ the k-shift increases.

Surface roughness has in general a Gaussian distribution described by the ζ rms depth and the ξ mean correlation length of the irregularities [4]. Although the value of ζ and ξ can not be calculated in advance for a given surface, it can be obtained by fitting theoretical curves to experimental data [3,4,5]. The size and distribution of irregularities depend first on the quality of substrates and also on film thicknesses within the MOM structure furthermore on evaporation parameters. Therefore they vary slightly from sample to sample.

There are three possible sources of light emission from a MOM diode [6], namely

/i/ fluctuations in the tunnel current coupled directly to the field of light; this light is p-polarized and consists of photons with $h\nu \lesssim E_c = eU$ energy where U is the bias on the diode,

/ii/ the high momentum surface plasmons excited in the junction region being transferred into unpolarized photons by scattering on $\xi = 3$-5 nm surface irregularities, and

/iii/ the top electrode-vacuum interface plasmons /"fast mode"SPO/ coupled by $\xi \sim 500$ nm periodical surface corrugations to the EM field of light.

In the last two cases the emission of photons with $h\nu \leq E_d$ is expected where E_d is the d-absorbtion edge of the anode metal since the internal damping of plasmons in the $\hbar\omega > E_d$ range is too high.

This model seems to describe well the experimental data for Al-Al_2O_3-Ag diodes. It is believed, however, that the observed unpolarized background radiation originates rather from the "fast mode" SPO, scattered by $\xi \sim 100 - 500$ nm statistical surface roughness than from /ii/ [2].

More recently it was concluded from experiments on smooth substrate Al-Al_2O_3-Au diodes that in Au the /i/ type radiation can only be seen [7]. Since this significant difference between Au and Ag was considered by us to be highly improbable, diodes with Au top electrode were studied using the technique described in [2].

The Al-Al_2O_3-Au sturctures were prepared by vacuum evaporation on a $(1\bar{1}02)$ orientation sapphire single crystal substrate, supporting an ion-etched holographic grating with a=555 nm periodicity and $h \sim 6$ nm depth of the grooves. The value of h was evaluated from the intensity of the first diffraction maximum of a He-Ne laser beam [8]. The film thicknesses were measured to be 50 nm for Al, 2.5-3 nm for Al_2O_3 and 35 nm for Au. Light emission was observed from the structures if U>2 V DC bias was applied and the Au electrode served as the anode. The maximal bias used in the experiments was 3.3 V when typically 10 mA tunnel current was measured.

The spectral and angular distribution of the emitted light was measured at room temperature as described in [2]. The $\omega(k)$ dispersion relation of SPO was calculated from the peaks observed in the angular distribution, making use of the momentum conservation law for the plasmon-photon system.

Similar dispersion relations were obtained as those for Ag in [3]. There is a small but significant shift towards higher k-values, compared to the dispersion relation calculated from the optical constants of [9]. This can be explained by the influence of surface roughness of the Au-vacuum interface /Fig.1/. By fitting the experimental data to the theory [4,5] ξ= 200 nm and ζ= 3.5 nm was obtained for the roughness parameters in good agreement with Ag data on the same substrate [3].

It is seen in Fig.2 that the intensity of plasmon peaks cuts off at $E_d \sim 2.45$ eV. Above this energy plasmons can practically not exist due to the d-band absorbtion. The peak in the plasmon intensity curve of Fig.2 at E = 2.1 eV is the result of the incoherent superposition of the n=+1 and n=-1 plasmon branches and comes from plasmons satisfying the [2k]=2G Bragg condition. If the data of Fig.2 are compared with those for Ag as given in Fig.4 of [3] it can be concluded that the intensity of the p-polarized background radiation /I_b/ in the whole spectral region, and that coming from SPO /I_{pl}/ up to 2 eV, are nearly the same in both cases. This indicates that both in Au and Ag similar processes are responsible for the radiation.

The radiation, emitted into the direction normal to the surface /Θ=0/, consists of an unpolarized component ranging up to E_c and a p-polarized SPO peak determined by the lattice constant of the grating /Fig.3/. The position of this latter peak does not depend on the bias while the maximum of the background is shifted towards higher energies if the bias is increased as seen from the s-polarized curves of Fig.3. The form of this background curve is similar to that of /i/ calculated in [6] but it is not even partly polarized.

It is observed that for both components the quantum efficiency of radiation decreases if U is increased /Fig.3/. This may be due to the change of the mechanism of conduction, e.g.from tunneling to conduction in fibers at higher biases and therefore at higher currents.

To sum up we may conclude that the mechanism of light emission in Al-Al_2O_3-Au diodes is similar to that in Al-Al_2O_3-Ag diodes hence it can be described by three processes of which the smallest one is probably /i/. The second processes can be the scattering of the anode-vacuum SPO on statistically distributed roughnesses with a correlation length in the order of 100 nm /obtained from the dispersion curve fit parameters and simultaneously from electronmicrographs /and the third is /iii/. It is worthwhile to mention that in preliminary experiments Al-Al_2O_3-Cu diodes gave similar results as Au topped diodes and the SPO dispersion relation could be determined up to 2.1 eV, the d-absorption edge of Cu. Therefore it can be concluded that the light emission mechanism of all tunnel diodes with noble metal anode is qualitatively the same.

The authors are indebted to Dr. V.A.Sychugov / Lebedev Institute, Moscow / for preparing the holographic gratings.

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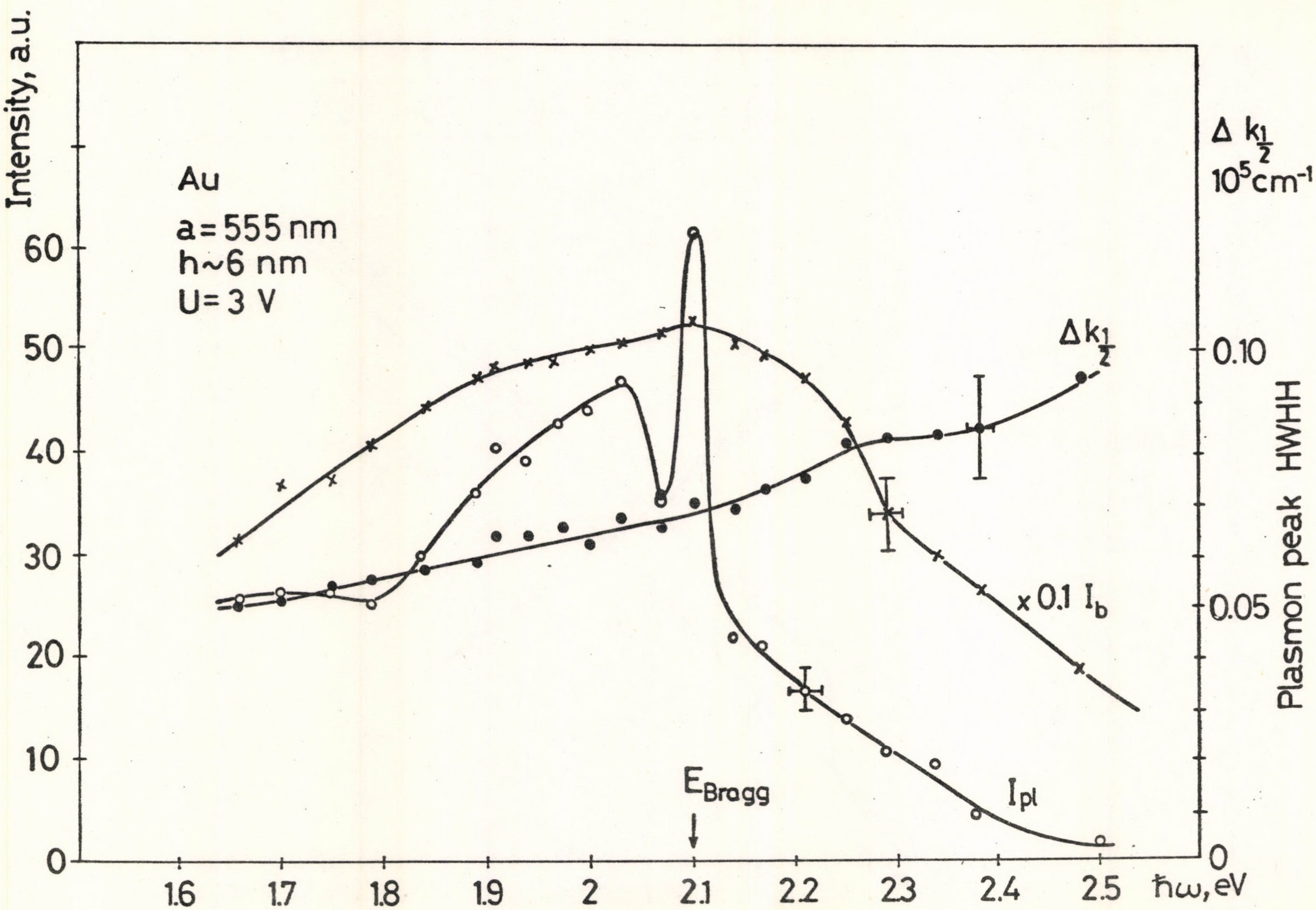
Intensity, a.u.
Au
a=555 nm
h~6 nm
U=3 V
60
50
40
30
20
10
0
1.6
1.7
1.8
1.9
2
2.1
2.2
2.3
2.4
2.5
ħω, eV
E_Bragg
Δk_{1/2}
10^5 cm^-1
0.10
0.05
0
Plasmon peak HWHH
x 0.1 I_b
I_pl

Fig. 2.

region being transferred into unpolarized photons by scattering on ξ = 3-5 nm surface irregularities, and

/iii/ the top electrode-vacuum interface plasmons /"fast mode"SPO/ coupled by $\xi \sim$ 500 nm periodical surface corrugations to the EM field of light.

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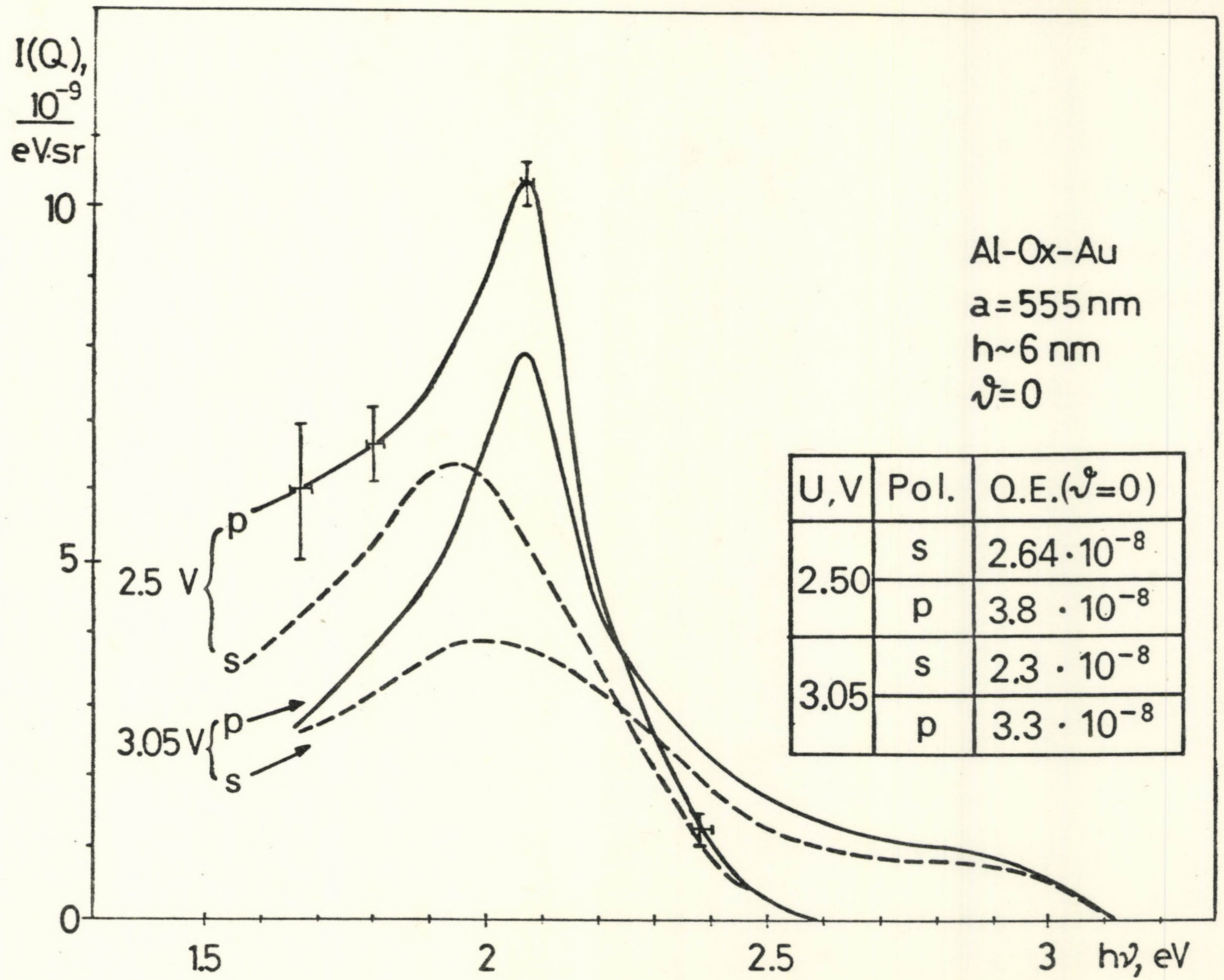


U, V	Pol.	Q.E. (ϑ=0)
2.50	s	$2.64 \cdot 10^{-8}$
	p	$3.8 \cdot 10^{-8}$
3.05	s	$2.3 \cdot 10^{-8}$
	p	$3.3 \cdot 10^{-8}$

Fig. 3.

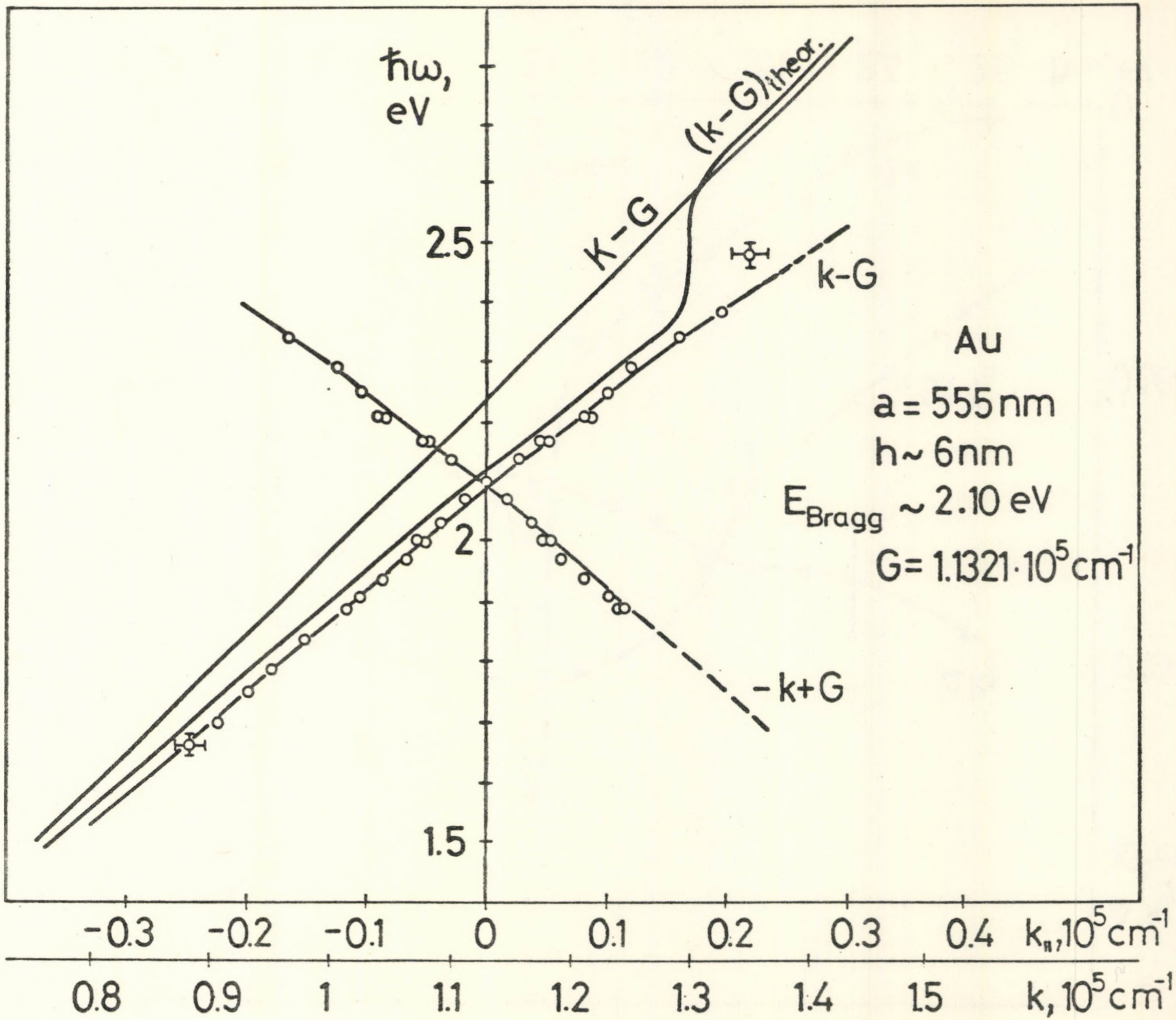
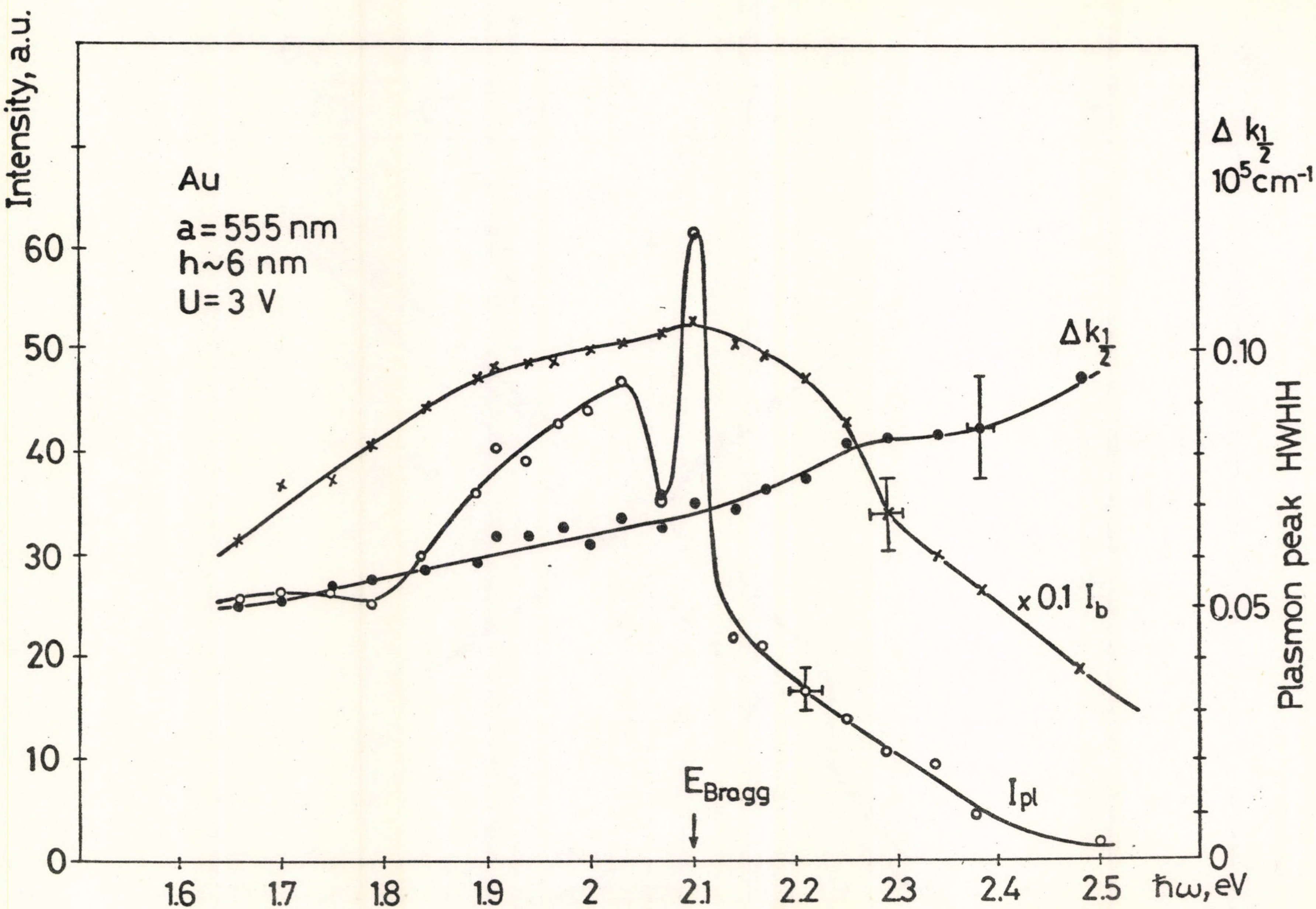
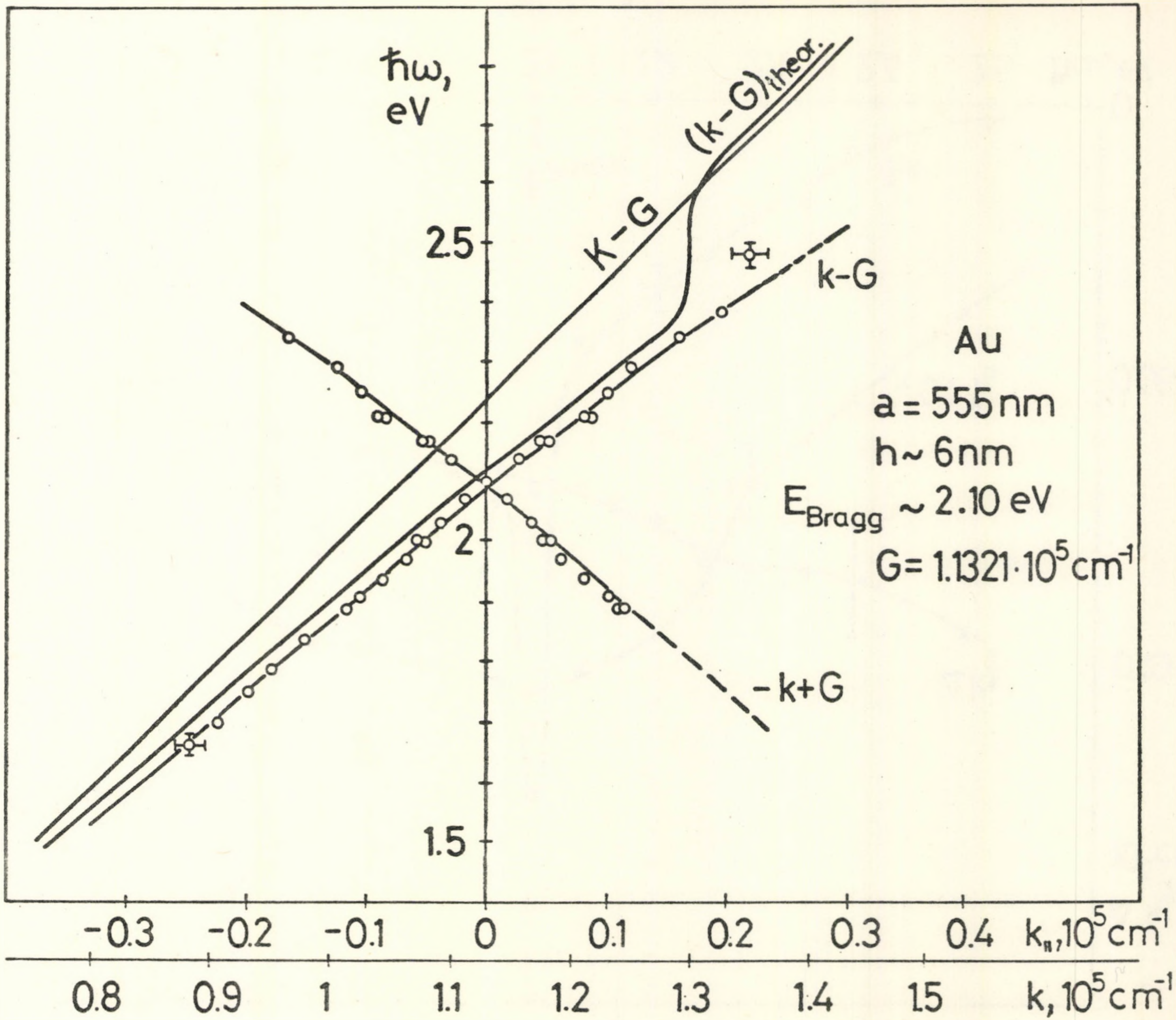

ħω, eV
2.5
2
1.5
(k-G)theor.
K-G
k-G
-k+G
Au
a = 555 nm
h ~ 6 nm
E_Bragg ~ 2.10 eV
G = 1.1321·10^5 cm^-1
-0.3 -0.2 -0.1 0 0.1 0.2 0.3 0.4
k‖, 10^5 cm^-1
0.8 0.9 1 1.1 1.2 1.3 1.4 1.5
k, 10^5 cm^-1

Fig. 1.



Intensity, a.u.
Au
a = 555 nm
h~6 nm
U = 3 V
$\Delta k_{\frac{1}{2}}$
$10^5 cm^{-1}$
Plasmon peak HWHH
x 0.1 I_b
I_{pl}
E_{Bragg}
60
50
40
30
20
10
0
0.10
0.05
0
1.6
1.7
1.8
1.9
2
2.1
2.2
2.3
2.4
2.5
ħω, eV

Fig. 2.



ħω, eV
2.5
2
1.5
K-G
(k-G)theor.
k-G
-k+G
Au
a = 555 nm
h ~ 6 nm
E_Bragg ~ 2.10 eV
G = 1.1321·10^5 cm^-1
-0.3
-0.2
-0.1
0
0.1
0.2
0.3
0.4
k∥, 10^5 cm^-1
0.8
0.9
1
1.1
1.2
1.3
1.4
1.5
k, 10^5 cm^-1

Fig. 1.

FIGURE CAPTIONS

Fig.1. SPO dispersion relation at the Au-vacuum interface. K is the light line, $k = k_{\parallel} + G$, where $G = 2\pi/a$ and $k_{\parallel} = K \sin\theta$. The full line through the experimental points is the result of a theoretical fit with roughness parameters $\xi = 200$ nm and $\zeta = 3.5$ nm. Bias voltage U = 3 V.

Fig.2. Plasmon peak half width, plasmon peak area $/I_{pl}/$ and p-polarized background intensity $/I_b/$ integrated in the equatorial plane for a space quarter on the n=-1 plasmon dispersion branch of Fig.1. Intensities are normalized to unit tunnel current.

Fig.3. Spectral distribution of the emitted light from a tunnel diode with Au anode at different biases. I/Q/ is the quantum efficiency of radiation in unit solid angle and energy interval. Quantum efficiencies of the emission into unit solid angle around $\theta = 0$ are $2.6.10^{-8}$ and $3.8.10^{-8}$ photon /electron/ sr for s and p-polarized radiation, respectively.

REFERENCES

[1.] J.R.Kirtley, T.N.Theis and J.C.Tsang, Appl.Phys.Lett. 37 /1980/435.

[2.] N.Kroo, Zs.Szentirmay and J.Félszerfalvi, Phys.Lett. 81A /1981/ 399.

[3.] N.Kroo, Zs.Szentirmay and J.Félszerfalvi, Report KFKI-1981-68 /submitted to Physics Letters A/.

[4.] J.A.Bush, D.K.Cohen, K.D.Scherkoske and S.O.Sari, JOSA 70 /1980/ 1020.

[5.] F.Toigo, A.Marvin, V.Celli and N.R.Hill, Phys.Rev. B15 /1977/ 5618.

[6.] B.Laks and D.L.Mills, Phys.Rev. B20 /1979/ 4962; Phys.Rev. B22 /1980/ 5723.

[7.] K.Parvin and W.Parker, Solid State Comm. 37 /1981/ 629.

[8.] W.J.Tomlinson and H.P.Weber, JOSA 63 /1973/685.

[9.] H.J.Hagemenn, W.Gudat and C.Kůnz, DESY Report N.SR-74/7, 1974.

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